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GLASS FIBERS AND A METHOD FOR THE MANUFACTURE OF THE
GLASS-FIBER-REINFORCED POLYPHENYLENE ETHER USING THESE

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[There are no amendments to this patent.]

Abstract

Objective

During the reinforcement of a polyphenylene ether resin (also including modified polyphenylene ether) with glass fibers, glass fibers treated with a special agent are used to increase the adherence between the polyphenylene ether (to be abbreviated as PPE hereafter) resin and glass fibers to obtain a molded

product with a remarkable improvement in the reinforcing effect of such glass fibers with respect to the PPE resin.

Constitution

Glass fibers containing more than 0.01% of a boron compound in the treating agent for glass fibers with respect to the weight of the glass fibers, as well as a method for the manufacture of glass-fiber-reinforced polyphenylene ether obtained by molding using these.

Claims

1. Glass fibers treated with an agent containing a boron compound.
2. A method for the manufacture of glass-fiber-reinforced polyphenylene ether molded using glass fibers treated with an agent containing a boron compound.

Detailed explanation of the invention

[0001]

Industrial application field

The present invention relates to glass fibers that can be used in glass-fiber-reinforced polyphenylene ether (to be abbreviated as FR-PPE hereafter) and to a method for the manufacture of the FR-PPE molded using these.

[0002]

Prior art

Conventionally, in the manufacture of glass fibers, a method is carried out in which glass is melted in a furnace, drawn from multiple small holes called bushings, applied with an appropriate treating agent via an applicator, stretched so that it is from a few microns to more than 10 microns, then dried. The glass-fiber treating agent used for the FR-PPE consists of a film forming agent with a synthetic resin as a major component, a coupling agent, a surfactant, a lubricant, an electrostatic inhibitor, etc. For the glass fibers treated with such a publicly known treating agent, in the case of kneading with PPE by an extrusion molding machine or an injection molding machine, the adhesiveness of the PPE and the glass composition is poor and the mechanical characteristics of the molded product are insufficient.

[0003]

Problems to be solved by the invention

The task to be solved by the present invention is to provide glass fibers with an increased adhesiveness between such glass fibers and the PPE resin, and to provide an improvement in the reinforcing effect of glass fibers with respect to the PPE resin. Furthermore, using the glass fibers mentioned previously, a method is provided for the manufacture of a molded product

having remarkably better physical properties than the conventional ones.

[0004]

Means to solve the problems

The present invention has been discovered as a result of the accumulation of zealous investigations in order to upgrade the reinforcing effect of glass fibers with FR-PPE. In other words, the present invention involves glass fibers characterized by the fact a boron compound is blended in a treating agent of such glass fibers. Furthermore, it is a method for the manufacture of FR-PPE molded using this. As the boron compounds mentioned in the present invention, sodium borate, boric acid, boric acid anhydride, ammonium borate, and sodium hydrogen borate can be mentioned. The specially preferred one is boric acid or boric acid anhydride. It is present at more than 0.001 wt% and less than 2 wt% with respect to the weight of the glass fibers. If the boron compound is present at less than 0.001 wt%, the adhesiveness with the PPE is not satisfactory. If it exceeds 2 wt% it is not economical. It is also acceptable that the treating agent referred to in the present invention be used together with a publicly known synthetic resin emulsion coupling agent, surfactant, etc. As the synthetic resin emulsions, those of polyvinyl acetate, unsaturated polyesters, polyethylene, polyurethane, epoxy resins, polystyrene, acrylic resins, or their copolymers and so on can be mentioned. Among these, one or more may be used in combinations. Furthermore, a variety of ordinary coupling agents added to the treating agents

of glass fibers include, for example, one or more coupling agents selected from vinyl tris (β -methoxyethoxy)silane, γ -methacryloxypropyltrimethoxysilane, γ -glycidoxypropyltrimethoxysilane, γ -aminopropyltriethoxysilane, methacrylate chromic chloride, N-B(N-vinylbenzylamino), γ -aminopropyltriethoxy silane hydrogen chloride, etc. It is also acceptable to add appropriately nonic or cationic surfactants, for example, one or more surfactants selected from polyoxyethylene alkyl ethers, polyoxyethylene alkylphenols, fatty acid sorbitan esters, alkyl quaternary ammonium salts, alkylamine salts, alkyl imidazoline derivatives, etc. Furthermore, it is also acceptable to use a lubricant, an antioxidant, an electrostatic inhibitor, or other additives. The compositions of the glass fibers of the present invention are glass for electrical insulation and ordinary E glass. In addition, C glass, A glass, T glass, and so on may also be appropriately used. The PPE that can be used in the present invention includes poly-2,6-dimethyl phenylene ether and modified polyphenylene ether. The modified polyphenylene ether includes the materials obtained by the blending and kneading of ABS, AS, and other polystyrene-type resins, as well as high-impact polystyrene, polybutadiene, and other diene-type rubber or nylon resins, and so on used as modifying agents in the poly-2,6-dimethyl phenylene ether mentioned previously. The FR-PPE of the present invention is obtained by the kneading and molding of the previously mentioned glass fibers and PPE by a publicly known extruding machine or injection molding machine. However, in these kneading methods, ordinary methods can be used without special restrictions. The blending amount of glass fibers in the molding material is from 5-50 wt%. Ordinarily,

15-30 wt% is appropriate. Furthermore, depending on the application, a flame retardant, filler, stabilizer, plasticizer, pigment, and so on can be blended.

[0005]

Application examples

Application examples are given in the following and the present invention will be explained in detail. However, the present invention is not to be restricted by these application examples in any way.

Application Examples 1 through 5

Treatment was carried out with the compositions shown in Table I as glass-fiber treating agents. The amounts showed the wt% with respect to the weight of the glass fibers.

Table I

処理剤組成 (1)	実施例 (2)				
	1	2	3	4	5
(3) ポリウレタンエマルジョン	1.0部 (10)	1.0部 (10)	1.0部 (10)	1.0部 (10)	
(4) γ-アミノプロピルトリエトキシラン	0.1部 (10)	0.1部 (10)	0.1部 (10)	0.1部 (10)	
(5) ホウ酸	0.01部 (10)	0.1部 (10)	1.0部 (10)		
(6) ホウ酸アンモニウム				0.1部 (10)	
(7) ホウ酸ナトリウム					0.1部 (10)
(8) 脱イオン水	残部 (11)	残部 (11)	残部 (11)	残部 (11)	残部 (11)

(9) *ポリウレタンエマルジョン：無黄変性ポリウレタンの水分散液

- Key: 1 Treating agent composition
 2 Application Example
 3 *Polyurethane emulsion
 4 γ-Aminopropyltriethoxysilane
 5 Boric acid
 6 Ammonium borate
 7 Sodium borate
 8 Deionized water
 9 *Polyurethane emulsion: Water-dispersed solution of non-yellowing polyurethane
 10 Part
 11 Balance

The treating agent mentioned previously was coated on 1600 glass filaments with a filament diameter of 13 μm obtained by melt-spinning using an applicator roll; these were wound on a rotary drum with a diameter of 300 mm. This strand wound body was afterwards cut to 3 mm with a cutting machine and dried with

a dryer at 130°C for 60 min to remove the water to obtain the treated glass fibers.

Table II

① 樹脂組成	A	B
② 日本GEプラスチック社製ノリル731J PPE樹脂一般タイプ	70部⑤	-
③ 日本GEプラスチック社製ノリルSEI PPE樹脂難燃タイプ	-	70部⑤
④ ガラス繊維	30部⑤	30部⑤

- Key: 1 Resin composition
 2 Noryl 731 J PPE resin, general type, manufactured by Japan GE Plastics Co.
 3 Noryl SEI PPE resin, difficulty combustible type, manufactured by Japan GE Plastics Co.
 4 Glass fibers
 5 Parts

Next, the resin composition shown in Table II was mixed in a blender, then extruded with a 30-mm-diameter biaxial extruder at a melt temperature of 290°C, forming pellets of FR-PPE with a pelletizer. These pellets were subjected to injection molding at a cylinder temperature of 300°C and a mold temperature of 80°C to obtain a molded product of FR-PPE. The evaluations of the molded products were carried out according to JIS K-7113, K-7203, and K-7110 to measure the tensile strength, bending

strength, and Izod impact strength. The adhesiveness between the glass fibers and the PPE resin was obtained by visual observation of the ruptured surface of the molded product after the impact test using SEM. The results are shown in Table III.

[0006]

Comparative example

Glass fibers and molded products were manufactured and evaluated by the same methods as those shown for Application Examples 1-5, except that no boric acid was added in Application Example 1. The evaluation results are shown in Table III.

[0007]

Effects of the present invention

According to the present invention, it is obvious that the adhesiveness between the glass fibers and PPE resin is good, and that the mechanical strength of the FR-PPE molded product is remarkably good.

Table III

	実施例①					比較例②	実施例①					比較例②
	1	2	3	4	5	1	1	2	3	4	5	1
樹脂組成③	Aの場合⑧						Bの場合⑨					
引張り強度④ (kg/cm ²)	1150	1200	1190	1200	1200	1100	830	950	930	920	980	780
曲げ強度⑤ (kg/cm ²)	1660	1750	1720	1780	1770	1520	1260	1350	1330	1330	1350	1150
アイゾット衝撃強度⑥ (kg·cm/cm)	14.8	15.5	15.5	15.8	15.5	13.7	7.2	8.5	8.8	8.6	8.5	6.6
*SEMによるガラス繊維とPPE 樹脂との界面の接着性の観察⑦	○	◎	◎	◎	◎	△	△	◎	◎	◎	◎	×

- Key: 1 Application Example
 2 Comparative Example
 3 Resin composition
 4 Tensile strength (kg/cm²)
 5 Bending strength (kg/cm²)
 6 Izod impact strength (kg·cm/cm)
 7 *Observation of adhesiveness of the rupture surface of glass fibers and PPE resin by SEM
 8 Case A
 9 Case B

Adhesiveness of glass fibers-PPE resin:

- | | |
|-------------------|---|
| Wetted very well | ◎ |
| Wetted well | ○ |
| Slightly wetted | △ |
| Not wetted at all | × |